

Application No. 09/720,068

Filed: March 8, 2001

Group Art Unit: 1754

REMARKS

Claims 1-18 were pending in the instant application. Claims 2, 6, 11, 15 and 17 have been amended herein. Accordingly, claims 1-18 will be pending upon entry of the amendments presented herein.

Support for the claim amendments can be found throughout the specification and claims as originally filed. Support for the claim amendments were prepared in response to the objections and rejections further explained below. No new matter has been added.

Amendments to and cancellation of the claims made herein were done solely to expedite prosecution of the present application, and are not to be construed as an acquiescence to any of the rejections/objections made in the instant Office Action or any Office Actions in the parent application. Applicant reserves the right to further prosecute the claims as originally filed or similar claims in this or one or more subsequent patent applications.

Oath/Declaration

The Declaration has been objected to for having an incorrect application number. A newly executed Declaration correcting the typographical error to the application number is submitted separately herewith.

Claim Objections

Claim 6 has been objected to for misspelling the word "oxidizing." Applicant has corrected the typographical error.

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Claim Rejections - 35 U.S.C. §112

Claims 2, 11, 15 and 17 are rejected under 35 U.S.C. §112, second paragraph for indefiniteness. In accordance with the Examiner's suggestions, claims 2, 11, 15 and 17 have been amended herein to appropriately recite the proper Markush language, thereby overcoming the foregoing rejection.

Claim Rejections - 35 U.S.C. §103

Claims 1-5, 8-12 and 14 are rejected under 35 U.S.C. §103(a) as being obvious over Fernholz et al. (U.S. Patent 4,092,267).

Applicant respectfully traverses the foregoing rejection.

The present invention is directed to a process for the regeneration of a catalyst, where the catalyst comprises at least one precious metal on an amorphous silica-alumina support. The catalyst is impregnated with an acid in liquid state, followed by reduction or oxidation of the impregnated catalyst at a temperature above 200°C.

Unlike the present invention, Fernholz et al. is directed to a process for regenerating a palladium carrier catalysts that are used for the oxacylation of olefins in the gaseous phase. The regeneration process includes treating the exhausted palladium carrier catalyst with a mixture consisting of a fatty acid that is liquid under normal conditions as a first component and, as a second component, nitric oxides, optionally with the addition of oxygen or oxygen-containing gases, nitric acid, peroxides, or peracids, drying the treated catalyst at a temperature below 90° and evaporating the fatty acid to a residual content of below 8 weight percent. (See Patent 4,092,267, column 3, lines 5-16, and claim 1, column 5, line 12, to column 6, line 3.) This regeneration process is particularly for the preparation of vinyl

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acetate from ethylene or of allyl acetate from propylene, acetic acid and oxygen. Fernholz et al. describes the preparation of palladium catalysts using all kinds of palladium salts, including chlorides and sulfates. When chloride or sulfate is used, the chlorine of the sulfate has to be removed from the impregnation solution. This is a method for the preparation of an original catalyst. The process of Fernholz et al. is distinguishable from the present invention such that no teaching or suggestion can be read with respect to the present invention.

The Examiner states that Fernholz et al.'s "palladium catalysts are impregnated with diluted HCl and are reduced with hydrogen gas at 200°C (col. 2, lines 53-55 and col. 3, lines 27-31)." (Office Action, bottom of page 3 to top of page 4.) The Examiner considers that one of ordinary skill in the art would have found it obvious, based on the teachings of Fernholz et al., "to impregnate a silica-alumina support having a palladium metal dispersed thereon with an acid and then to reduce the catalyst at a temperature above 200°C, because Fernholz et al. discloses a process for regenerating palladium catalysts, having aluminosilicate supports, wherein the palladium catalysts are impregnated with diluted HCl and are reduced with hydrogen gas at 200°C." (Office Action, page 4, 2<sup>nd</sup> paragraph.)

To the contrary of Examiner's view, any skilled person in the art would not consider the process described in column 2, lines 48-65, of the '267 patent as that of the process Fernholz et al. is using. In fact, the process described in column 2 that the Examiner is basing as that of Fernholz et al. process is to a known German process "Offenlegungsschrift" No. 2,420,374. Fernholz et al. describes this known process as being "disadvantageous on an industrial-technical scale due to the seven

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separate steps required for its execution." (The '267 patent, column 2, line 68, to column 3, line 2.) By its reference to this method of impregnation with diluted hydrochloric acid, Fernholz et al. does not imply using this step in its process. If anything, Fernholz et al. is teaching away from using this method, or any of the steps, as applied to the present invention since Fernholz et al. discourages the particular method steps to be used on an industrial technical scale. The present invention overcomes this challenge by providing a combination of steps for a proper re-dispersion of the precious metal catalyst, by impregnating the catalyst with an acid, followed by reduction or oxidation of the impregnated catalyst at a temperature above 200°C. The method as claimed provides a reactivated catalyst using a relatively simple process that can be used on an industrial technical scale.

Moreover, the Examiner has not provided any motivation to combine the one step in the method described in column 2, lines 50-65, with a hydrogen gas reduction at 200°C described in column 3, lines 27-31, to come up with claim 1 of the invention. "[I]denfication in the prior art of each individual part claimed is insufficient to defeat patentability of the whole claimed invention... Rather, to establish obviousness based on a combination of the elements disclosed in the prior art, there must be some motivation, suggestion or teaching of the desirability of making the specific combination that was made by the applicant." *In re Kotzab*, 55 U.S.P.Q.2d 1313, 1316 (Fed. Cir. 2000). No requisite motivation has been proffered to be able to come up with the claimed invention, especially since contradictory statements are made with respect to one of the steps the Examiner proposes that Fernholz et al. practices. Fernholz et al. is directed to the oxacylation of olefins and reasons that the steps used in the

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method of the known German process for regenerating palladium metal carrier catalysts is disadvantageous. No motivation can be gleaned from Fernholz et al. to come up with the particular steps in the process for the regeneration of a catalyst as claimed.

Furthermore, the Examiner considers that "it would have been obvious to one ordinary skill to reduce at a temperature above 200°C, because the temperature disclosed by Fernholz et al. is close enough that one skilled in the art would have expected it to have the same properties." (Office Action, page 4, 2<sup>nd</sup> paragraph.) Applicant respectfully disagrees. Plainly, Fernholz et al. advocates 200°C or less in its process. Process conditions can vastly change the resulting product. No motivation can be found in Fernholz et al. to use a reduction or oxidation temperature of greater than 200°C in a re-dispersion of a precious metal catalyst, by impregnating the catalyst with an acid. It would not be obvious to combine certain known steps that are considered disadvantageous with steps that Fernholz et al. practices. Fernholz et al. fails to teach or suggest the invention as claimed.

Claims 6, 13, 15 and 16 are rejected under 35 U.S.C. §103(a) as being unpatentable over Fernholz et al. as applied to claims 1-5, 8-12 and 14, and further in view of Berrebi (U.S. Patent 5,068,477) and Kojima et al. (U.S. Patent 5,391,527).

Applicant respectfully traverses the rejection.

As explained above, Fernholz et al. cannot make the present invention obvious. Berrebi and Kojima et al., either alone or in combination with Fernholz et al., would not cure the deficiencies found in Fernholz et al., the primary reference. The teachings of Berrebi, in and of itself, would not make the present invention obvious. Berrebi is directed to a process for reducing a refining

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catalyst before it is put into use. The catalyst in Berrebi is used in selective hydrogenation of a feed such as olefin, acetylenes, diolefins, etc. (see Abstract, '477 patent.) The method in Berrebi consists of impregnating a catalyst with an aqueous or organic solution of a compound that is a reducing agent, heating the catalyst for decomposing the reducing agent at a temperature between 100°C and 150°C and then drying the catalyst. In contrast, the present invention is directed to the regeneration of a catalyst, where the catalyst comprises at least one precious metal on an amorphous silica-alumina support, in which the process comprises impregnating the catalyst with an acid in liquid state and reducing or oxidizing the impregnated catalyst at a temperature above 200°C. The present method is a very simple process for regenerating a used catalyst, whereas the Berrebi method applies to reducing a refining catalyst before it is used. Additionally, the conditions used in Berrebi are distinguishable from the claimed process. The divergent teachings of Berrebi, alone or in combination with Fernholz et al., would not provide an ordinary skilled artisan the motivation nor the idea of regenerating a used catalyst in the process steps and conditions as claimed.

Similarly, Kojima et al. cannot cure the deficiencies found in either Fernholz et al. or Berrebi. Kojima et al. describes regenerating deactivated alkylation catalysts where the alkylate feedstock has been removed with hydrogen at temperatures in the range of 10°C to 300°C. Kojima et al. is distinguishable from the present invention, which is directed to the regeneration of a catalyst comprising at least one precious metal on an amorphous silica-alumina support, where the catalyst is impregnated with an acid in liquid state, followed by reduction or oxidation of the

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impregnated catalyst at a temperature above 200°C. Kojima et al. uses hydrogen gas (see U.S. Patent 5,391,527, claim 1, column 8, lines 20-21) and not an acid in liquid state. In fact, Kojima et al. emphasizes removing all liquid phases in the alkylation catalyst. With all of the deficiencies described in Fernholz et al., Kojima et al., either alone or in combination, would not make the present invention obvious; no motivating factor is apparent from the teachings of Kojima et al. None of the cited references, alone or in combination, teaches or suggests the invention as claimed herein. The cited references provide no requisite motivation to combine or the teaching to be able to come up with the claimed invention. Therefore, Applicant respectfully requests reconsideration and withdrawal of the foregoing rejection.

Claims 7, 17 and 18 are rejected under 35 U.S.C. §103(a) as being obvious over Fernholz et al. (U.S. Patent 4,092,267) in view of Berrebi (U.S. Patent 5,068,477) and Kojima et al. (U.S. Patent 5,391,527) as applied to claims 1-6 and 8-16 above, and further in view of Neuenfeldt et al. (U.S. Patent 5,695,634).

Applicant respectfully traverses the foregoing rejection.

Previous arguments made herein apply with respect to Fernholz et al., Berrebi and Kojima et al. Neuenfeldt et al. fails to cure the deficiencies found in Fernholz et al., in view of Berrebi and Kojima et al. Neuenfeldt et al.'s process of regenerating a catalyst for wastewater treatment containing organohalogens includes by first washing with acetone and then with hydrochloric acid at room temperature (see column 3, lines 22-24) or at a temperature of at least 500°C (see column 3, lines 27-29). Plainly, Neuenfeldt et al., in and of itself, is distinguishable from the present invention. While Neuenfeldt et al. may describe a support prepared by a sol-gel process, it is silent with respect

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to the particular combination of process steps for the regeneration of a precious metal catalyst. Neuenfeldt et al. fails to teach one of ordinary skill in the art to come up with a process for regenerating a precious metal on an amorphous silica-alumina support catalyst by impregnation with an acid in liquid state, followed by an oxidation or reduction at a temperature above 200°C. Moreover, Neuenfeldt et al. in combination with the other cited references would not lead one of ordinary skill in the art to come up with the present invention as claimed. No requisite motivation can be gleaned from the cited references based on the deficiencies described above, either alone or in combination, to render the present application obvious.



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CONCLUSION

Based on the foregoing, Applicant respectfully requests reconsideration of the pending claims and allowance of the application.

The Examiner is encouraged to telephone the undersigned attorney to discuss any matter that would expedite allowance of the present application.

Respectfully submitted, **FAX RECEIVED**

MARIUS VAARKAMP

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GROUP 1700

By: 

Stanley M. Schurgin  
Registration No. 20,979  
Attorney for Applicant(s)

WEINGARTEN, SCHURGIN,  
GAGNEBIN & LEOVICI LLP  
Ten Post Office Square  
Boston, MA 02109  
Telephone: (617) 542-2290  
Telecopier: (617) 451-0313

CSK/knr

Enclosure - A newly executed Declaration  
290873-1

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AMENDMENT TO THE CLAIMS

Claim 1. (Previously amended) Process for the regeneration of a catalyst, said catalyst comprising at least one precious metal on an amorphous silica-alumina support, in which process the catalyst is impregnated with an acid in liquid state, followed by reduction or oxidation of the impregnated catalyst at a temperature above 200°C.

B1  
Claim 2. (Currently amended) Process according to claim 1, wherein the precious metal is ~~at least one~~ selected from the group consisting of Pt, Pd, Au, Ir, Ru, Rh, Re, Os and Ag, preferably Pt and/or palladium.

Claim 3. (Previously amended) Process according to claim 1, wherein the degree of dispersion is increased after the regeneration.

Claim 4. (Previously amended) Process according to claim 1, wherein the acid impregnated catalyst is reduced in a flow of hydrogen gas.

Claim 5. (Previously amended) Process according to claim 1, wherein the acid impregnated catalyst is oxidised in a flow of dry (<0.1 vol. % of water) air, followed by reduction.

B2  
Claim 6. (Currently amended) Process according to claim 1, wherein the reduction and ~~and/or oxidising~~ oxidizing step are carried out at a temperature of between 250 and 600°C.

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Claim 7. (Previously amended) Process according to claim 1, wherein the silica-alumina support has been prepared using a sol-gel method.

Claim 8. (Previously amended) Process according to claim 1, wherein the support has an Si-Al atomic ratio of from 0.1 to 300.

Claim 9. (Previously amended) Process according to claim 1, wherein the catalyst has a precious metal content of from 0.01 to 5 wt.%, calculated on the basis of the weight of reduced catalyst.

Claim 10. (Previously amended) Process according to claim 1, wherein the catalyst is impregnated with an aqueous solution of the acid.

*B3*  
Claim 11. (Currently amended) Process according to claim 1, wherein the acid is selected from the group consisting of HCl, H<sub>3</sub>PO<sub>4</sub>, H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub>, HBr and combinations thereof.

Claim 12. (Previously amended) Process according to claim 1, wherein the amount of acid calculated on the basis of a ratio equivalents of acid to atoms of precious metal is between 0.1 and 100, preferably between 0.5 and 10.

Claim 13. (Previously amended) Process according to claim 1, wherein prior to the impregnation, carbonaceous deposits on the catalyst are burned off.

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Claim 14. (Previously amended) Process according to claim 1, wherein the regeneration is carried out in a reactor, separate from the reactor in which the catalyst is used.

B<sup>4</sup>  
Claim 15. (Currently amended) Process according to claim 1, wherein the catalyst is used catalyst from a process inselected from the group consisting of hydrogenation, hydro-isomerisation, hydro-desulfurisation, hydrodewaxing and catalytic reforming.

Claim 16. (Previously amended) Process for hydrogenation, hydro-isomerisation, hydro-desulfurisation or hydrodewaxing, comprising treating the feedstock in the presence of a catalyst that has been regenerated using the process of claim 1.

B<sup>5</sup>  
Claim 17. (Currently amended) Process according to claim 2, wherein:

the degree of dispersion is increased after the regeneration;  
the acid impregnated catalyst is reduced in a flow of hydrogen gas;

the acid impregnated catalyst is oxidised in a flow of dry (<0.1 vol.% of water) air, followed by reduction;

the reduction and or oxidising step are carried out at a temperature of between 250 and 600°C;

the silica-alumina support has been prepared using a sol-gel method;

the support has an Si-Al atomic ratio of from 0.1 to 300;

the catalyst has a precious metal content of from 0.01 to 5 wt.%, calculated on the basis of the weight of reduced catalyst;

the catalyst is impregnated with an aqueous solution of the acid;

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the acid is selected from the group consisting of HCl, H<sub>3</sub>PO<sub>4</sub>, H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub>, HBr and combinations thereof;

the amount of acid calculated on the basis of a ratio of equivalents of acid to atoms of precious metal is between 0.1 and 100, preferably between 0.5 and 10;

prior to the impregnation, carbonaceous deposits on the catalyst are burned off;

the regeneration is carried out in a reactor, separate from the reactor in which the catalyst is used; and

the catalyst is a used catalyst from a process in-selected from the group consisting of hydrogenation, hydro-isomerisation, hydro-desulfurisation, hydrodewaxing and catalytic reforming.

Claim 18. (Previously added) Process for hydrogenation, hydro-isomerisation, hydro-desulfurisation or hydrodewaxing, comprising treating the feedstock in the presence of a catalyst that has been regenerated using the process of claim 17.